Nobel Lecture: Genesis and applications of attosecond pulse trains*

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I. INTRODUCTION

Attosecond pulses have come of age with the 21st century and are now routinely used in dynamical studies on the timescale of electron motion in atoms (the atomic unit of time is 24 as). Both their generation and characterization have made great progress over the last decade or so, with the current state of the art characterized by 53 ± 6 as (Li *et al.*, 2017) and 43 ± 1 as pulses with a central energy of 100–150 eV (Gaumnitz *et al.*, 2017).

Up to the end of the 1990s, laser pulses were limited to femtoseconds. The chirped pulse amplification of Strickland and Mourou (1985) allowed intense pulses, but the infrared wavelength was restricting the pulses to at least one optical cycle (the wavelength of 800 nm of titanium:sapphire corresponds to a period of 2.7 fs). On the other hand, since the early 1990s it had been known that high harmonic generation was, in theory, a possible road for breaking the "femtosecond barrier" (Farkas and Toth, 1992). Several groups in the world were following that road, but it was only at the turn of the millennium that a method to determine the phases of a group of harmonics was demonstrated (Paul *et al.*, 2001) and named RABBITT by Muller (2002). The era of attophysics was born and the huge number of publications which followed the experimental availability of either attosecond pulse trains (APTs) or isolated attosecond pulses (IAPs) (Hentschel *et al.*, 2001) is a witness to the success of both techniques in atomic, molecular, and solid-state physics.

In this review, I wish to recall the first experimental determination of high harmonic spectral phase, and hence the first documented attosecond pulse train. Next, I will briefly review two applications of APTs: the photoionization delay near a Cooper minimum (Schoun *et al.*, 2014) and the nonsequential double ionization in the time domain, both recent experiments at Ohio State (Piper *et al.*, 2024).

II. FIRST PART

A. Prehistory

It is easy to realize that attosecond light pulses must have an optical period short enough so that at least one cycle is contained in the pulse. For, say, 100 as, the wavelength must be shorter than 30 nm (harmonic 27 of a Ti:sapphire laser at 800 nm). This implies that attosecond pulses propagate in vacuum and that, consequently, attosecond beam lines include steel tubing, pumps, etc. From that point of view, the attosecond regime is much more demanding than the femtosecond. Some attempts to generate a bandwidth large enough around an extreme ultraviolet (XUV) central wavelength. By the mid 1980s the discovery of the generation of harmonics of large orders at Chicago (McPherson *et al.*, 1987) and Saclay (Ferray *et al.*, 1988) was the first step on a long road to the APT. Anne L'Huillier, at Saclay and later in Lund, led research of several decades on high harmonics and applications of APT which was awarded by the 2023 Nobel Prize in Physics. One idea was suggested by Farkas and Toth (1992), to use high harmonics to synthesize subfemtosecond pulses. By Fourier transform, the spectrum of a series of N odd harmonics distant by twice the driving laser frequency corresponds in the time domain, a series of pulses separated by half the laser period, each pulse with a duration proportional to $1/2N$, assuming the spectral phase a linear function of frequency, or zero group delay (Fig. 1). It took almost ten years to implement a method to measure that phase and confirm Farkas and Toth. One of the reasons for this delay was perhaps a calculation by Anne L'Huillier herself (Antoine, L'Huillier, and Lewenstein, 1996) of the phases versus order which appeared to show that, due to an interference, the phases looked random, jumping by several 2π from an order to the next (Fig. 2). We will come back to this

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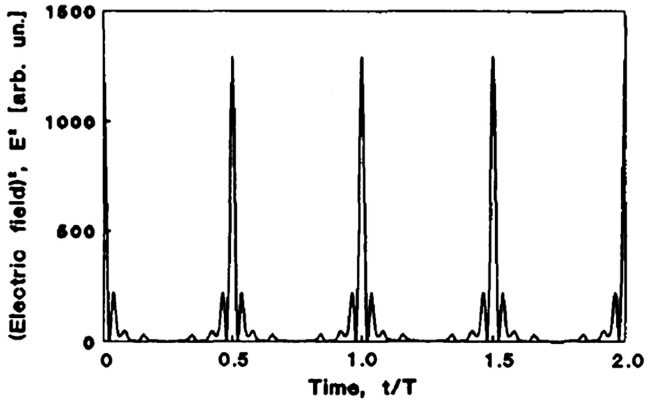

FIG. 1. The time profile of the APT, assuming the harmonics phase locked. From Farkas and Toth, 1992.

point later. A similar idea had been based on stimulated Raman scattering, but the bandwidth fell short of breaking the femtosecond barrier (Sokolov *et al.*, 2001). Although similar, this scheme had the handicap of the small spectral interval between the Raman sidebands. Eventually, the harmonics solution prevailed, despite their own handicap of a weak intensity which forbade direct nonlinear transitions. Now, these nonlinear processes are necessary to determine the phase, as a linear process is equivalent to measuring the power spectrum and hence contains no information about the phase. As we will see, this can be solved by mixing the weak harmonics beam with a laser strong beam. Let's just stress at this point that the phases are the property that determine whether the harmonic field shows strong amplitude modulation and makes possible the APT [see Antoine, L'Huillier, and Lewenstein (1996)].

B. Theories of HHG

In this section I briefly summarize the single-atom theories of high harmonics generation (HHG). For theories including propagation effects the reader is referred to L'Huillier, Schafer, and Kulander (1991). Harmonic generation is a nonlinear process in which an atom absorbs q photons of energy $\hbar\omega$ from a laser beam and emits one photon $\hbar\Omega$ such that $q\hbar\omega = \hbar\Omega$. In an atom, due to the symmetry of the potential, q must be odd (Fig. 3). For low values of q one can in principle calculate the rate of the process by time dependent perturbation theory. However, the experiment made rapidly obvious that q was large and a nonperturbative theory was required (L'Huillier and Balcou, 1993). Corkum (1993), Kulander, Schafer, and Krause (1993), and Schafer *et al.* (1993) proposed a solution by treating the problem classically for the part of the photoelectron motion in the continuum. This could even be simplified by neglecting the atomic potential in the Newton equation of motion. In that approximation the photoelectron is accelerated and brought back to the nucleus by the field where it converts its kinetic energy into photon energy (Fig. 4). This is the now well-known three-step model of strong field harmonic generation. The transition rate from the ground state to the continuum was calculated assuming a tunnel ionization (Ammosov, Delone, and Krainov, 1986). The "three-step model" approach was immediately successful by predicting the highest energy emitted (the "cutoff") as the highest kinetic energy (Corkum, 1993; Schafer *et al.*, 1993). The cutoff energy depends on the intensity and the ionization potential as $\hbar\Omega_{co} = 3.17\ U_p + IP$ where $U_p = I/4\omega^2$, the ponderomotive energy in atomic units, and IP is the atomic ionization energy. One of the consequences of the approach is

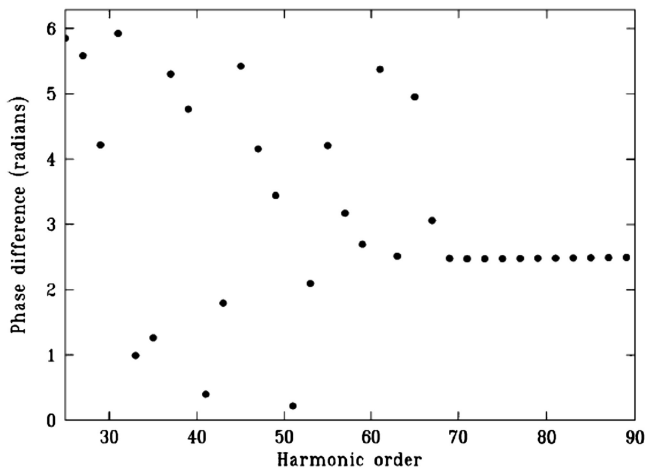

FIG. 2. Phases vs order according to the calculation of Antoine, L'Huillier, and Lewenstein (1996). Is the harmonic generation the solution?

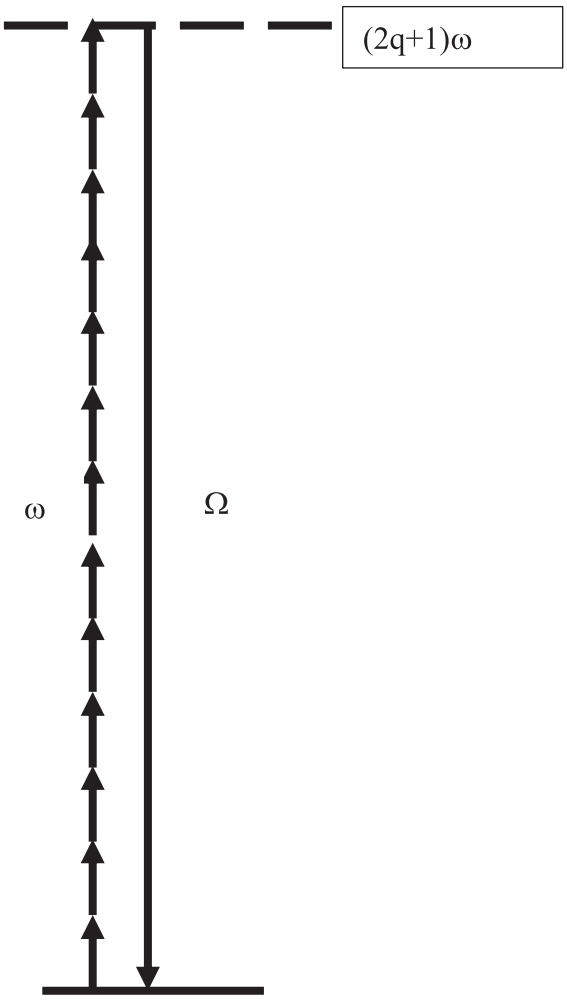

FIG. 3. The photon picture of harmonic generation.

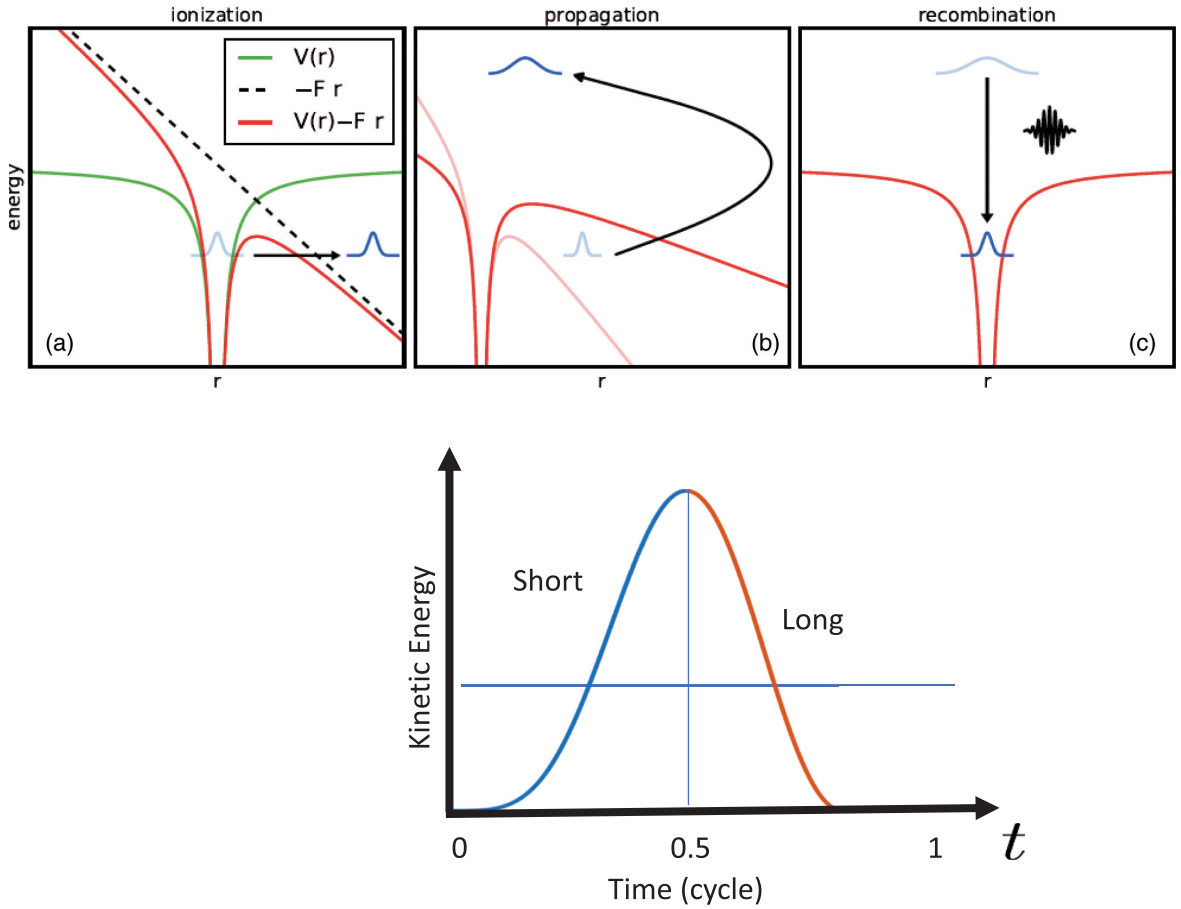

FIG. 4. Top panels: schematic of the three-step model. From Schoun, 2015. Bottom panel: photoelectron kinetic energy vs time. To a given energy (horizontal line) two trajectories (short and long) contribute. The maximum kinetic energy (cutoff) occurs close to a half cycle. From Kazamias and Balcou, 2004.

that for a given emitted energy (assumed to be an odd harmonic) there are two possible ionization times. The two electron trajectories starting and returning to the nucleus correspond to quantum mechanical transition amplitudes which interfere. This was readily noted in the first quantum theory (Lewenstein *et al.*, 1994) in a model atom, as the origin of oscillations of the harmonic amplitude when the harmonic order belongs to the "plateau," the region where all orders have a similar intensity, a clear nonperturbative signature. The two electron trajectories were dubbed "long" and "short." The long one starts earlier, closer to the peak of the field, and returns after the short one. Because of its starting time at higher laser field the long trajectory amplitude should be dominant. It's not, in practice because the short trajectory is selected. This important point will be taken in more details when discussing the results of the RABBITT measurement. By the end of the 1990s the theory of HHG was well understood and the basic result of Antoine, L'Huillier, and Lewenstein (1996) was still holding in front of the proposition of Farkas and Toth (1992). The need of a direct measurement of the harmonic phase was a clear necessity. Fortunately, the theoretical group at Pierre and Marie Curie University was developing the theory of such a measurement. We will first recall the process of above-threshold ionization (ATI) which is an important stage of that theory.

C. ATI

The Einstein photoeffect law of 1905 had to be deeply modified due to the advent of the laser beginning of the 1960s. First, due to multiphoton ionization. If an atom needs to absorb N_0 photons to be ionized with $N_0\hbar\omega \geq IP + U_p$ the kinetic energy of the photoelectron is $KE = N_o\hbar\omega - IP - U_p$ instead of $\hbar\omega - IP$ in the case of single-photon ionization, the only one known at Einstein's time (here U_p takes into account the increase of the ionization potential due to the ac-Stark shift). This was a working assumption until the experiment

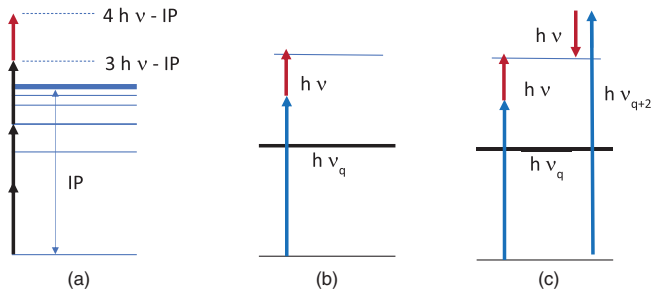

FIG. 5. Above-threshold ionization (ATI). One color (a), two color (b), and three color (c): two consecutive harmonics and a driving laser.

that measured the kinetic energy of the photoelectron for the first time (Agostini *et al.*, 1979). A surprise came with that experiment which showed that in a six-photon ionization xenon atom could absorb seven photons, the extra energy of one photon being converted into an excess of kinetic energy of the same amount. Following experiments confirmed that the complete photoeffect law reads $KE=(N_0+S)\hbar\omega-IP-U_p$ with S an integer number which can be very large. This process was baptized ATI [Fig. 5(a)]. Using XUV radiation from an attosecond pulse, single-photon ionization by a harmonic with an intensity larger than the ionization potential (the Stark shift is negligible) could make the first step of an ATI process. As a two-photon process, its probability is proportional to the square of the intensity, and it could be used to measure the spectral phase of the radiation. Unfortunately, due to the weak intensity of the harmonic radiation, driving a two-photon transition is out of the question, but by mixing it with a strong laser it can be done. In that perspective, the photoelectron results from the XUV + laser ATI process. Suppose a laser with photons ω, a harmonic with photons $(2q-1)\omega > IP$, then the minimum order ATI, harmonic + laser would yield a photoelectron of energy $E_{sb}=(2q-1)\omega+\omega-IP=2q\omega-IP$, where E_{sb} stands for the energy of the sideband. The harmonic of the next order would lead to $E_{sb}=(2q+1)\omega-\omega$, where this time a laser photon is emitted (Fig. 5).

D. Three-color ATI

It was the goal of the Veniard, Taieb, and Maquet (1996) calculation to evaluate the photoionization spectrum in the case of two harmonics of consecutive orders mixed to the laser

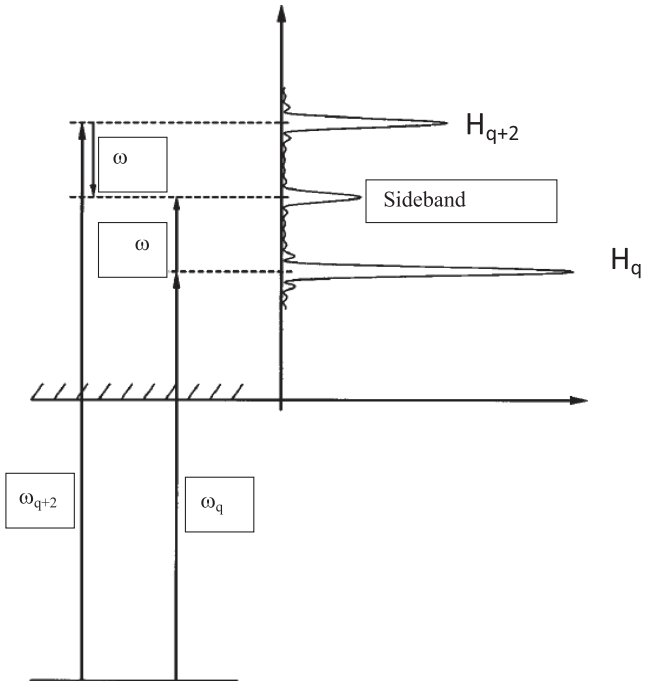

FIG. 6. Transitions involved in the phase measurement. The right part of the figure illustrates the photoelectron spectrum without laser (the two harmonic lines) and with it (the sideband). The two ATI transitions ending on the same state interfere. From Veniard, Taieb, and Maquet, 1996.

that generates them [Fig. 5(c)]. Besides the two direct one-photon amplitudes which determine the two basic lines in the spectrum there are two, two-photon, two-color (ATI) amplitudes ending in the same state called the sideband (Fig. 6). If the amplitudes are designated by M^{+}_{2q-1} and M^{-}_{2q+1} respectively for the lowest and the highest order, the sideband amplitude can be shown to be

$$S_q = C_q + |M^{+}_{2q-1}||M^{-}_{2q+1}|\cos(-2\omega\tau+\phi+\phi^{at}). \quad (1)$$

In Eq. (1) ω is the driving laser angular frequency, τ is a time delay between the harmonic and laser beams, the + and − signs indicate that the laser photon in the ATI transition is absorbed or emitted, respectively, ϕ is the sideband phase and ϕ^{at}, the atomic phase, a correction that must be calculated at this point. C_q contains no information on the phase. ϕ^{at}, dubbed the "atomic phase," depends on the atom being ionized. It has been calculated in various targets (He, Ne, Ar) at 800 nm the Ti:sapphire wavelength and is usually small. It can be considered a correction of the harmonic phase. Typically, this correction is only a small fraction of radians while the harmonic phase is of the order of 1 rad. This "correction" had however a great career under the name of "photoionization delay" [see Dahlström, L'Huillier, and Maquet (2012) for a tutorial]. In a model developed by Dahlström *et al.* (2013) the atomic phase delay can be split into the sum of two terms, the *Wigner-like delay* τ_η and the *continuum-continuum delay* τ_{cc}. For example, at a kinetic energy of 10 eV and 800 nm the first one is −70 as and the second −110 as (Schoun, 2015). The laser wavelength dependence is entirely contained in τ_{cc} and has a simple analytical expression, while the species dependence is contained in τ_η. Since the total delay is due to the argument of the two-photon matrix element $\arg(M^{+*}_{2q-1}M^{-}_{2q+1})$, the physical interpretation of τ_η and τ_{cc} is clear: it is the group delay for the single-photon ionization and the additional delay which occurs from the laser contribution to the two-photon ionization process. τ_η can be measured only in the two-photon process and cannot be measured independently of τ_{cc} (Schoun, 2015).

To conclude this paragraph, let us mention that the photoionization delays led to one of the first, if not the first, report on measured zeptosecond (10^{-21} s) times, as the delay found in the case of a H_2 molecule between the ionization times of the two electrons on each side of the molecule (Grundmann *et al.*, 2020). The delay was interpreted as the time the photon takes to cross the molecule. Let us stress also that the photoionization delay in the Véniard *et al.* calculation is linked to the ATI process through the argument of the two-photon matrix element which had been extensively studied during the preceding decades.

E. The RABBITT experiment

In this section I will recall the setup and the results of the first measurement of an APT. In modern attosecond beam lines it is usual to split the driving laser into two beams in a Mach-Zehnder interferometer. The first "historical" setup (Paul *et al.*, 2001), though, was much more compact and simpler, thanks to the ingenuity of Muller. A circular mask first transforms the incoming Gaussian laser profile into an annular beam which is normally focused into a first argon jet to generate the harmonics whose phases are to be determined.

Inside the jet the beam profile is Gaussian again, as if the mask did not exist (except for some energy missing). At some distance from the jet, the beam profile becomes annular again and a pinhole of suitable diameters is set to block most of the laser beam. The harmonic beam normally propagates through the central part of the pinhole and is directed to a second atomic jet of argon in which it creates photoelectrons which are subsequently analyzed in a magnetic bottle time-of-flight spectrometer [see Paul *et al.* (2001)]. The second laser beam (the "dressing beam") is created by a small aperture in the mask and propagates also along the axis. It can be delayed with respect to the first one, by a special couple of glass windows whose orientation determines an attosecond delay (this artifact is now visible at the Nobel Museum!). The plates were aligned parallel to each other with a precision of 0.01°. The plate responsible for the delay of the central part was first rotated by hand, then by a computer-controlled rotation stage which could make steps of 0.005°. Very small delays could be achieved (1 fs/ deg) close to normal incidence.

The dressing beam and harmonic beam are propagating together and are both focused by the same spherical mirror into the second atomic jet where it creates the sidebands whose amplitude versus delay determine the total harmonic phase $\phi + \phi^{at}$. Harmonics above argon ionization threshold were 11, 13, 15, 17, and 19. The first data obtained after normalizing to the total counts, to minimize the effect of the laser intensity fluctuations, are shown in Fig. 7. The superposition of the

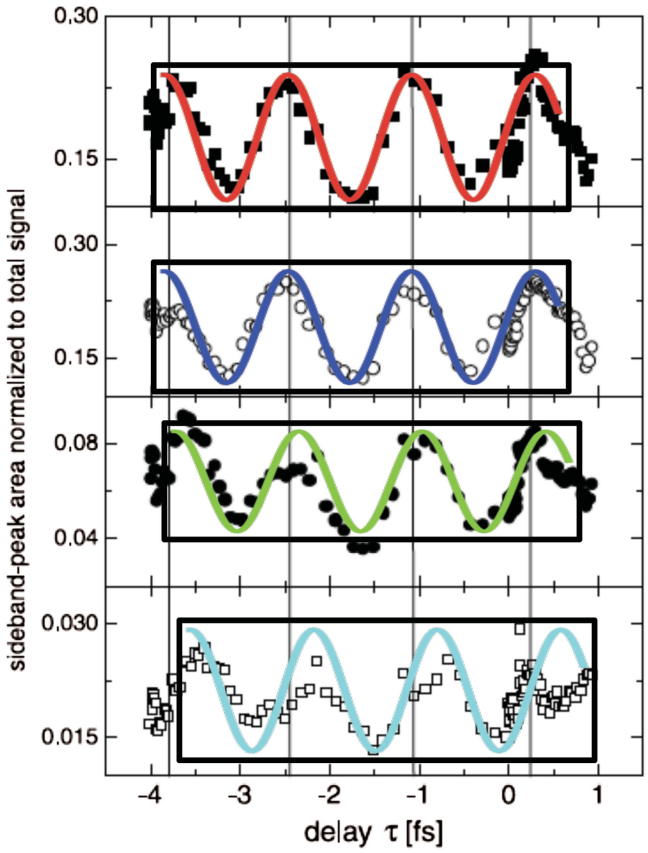

FIG. 7. Phases vs delay. The main difficulty with the experiment was the laser intensity fluctuations, which were partially removed by normalizing the signal to the total count. From Paul *et al.*, 2001.

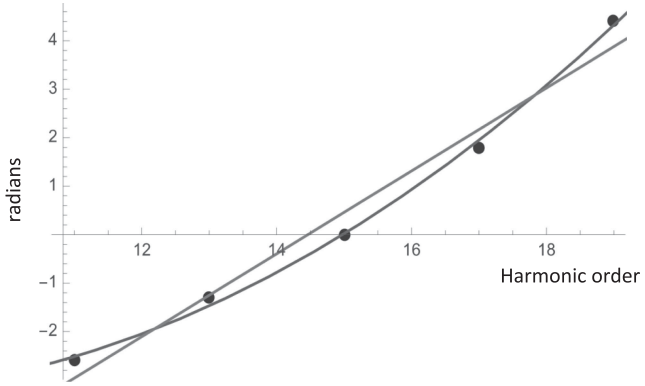

FIG. 8. Phases of harmonics 11 to 19 (dots) as measured by RABBITT. Together with the amplitudes, they determine the APT of Fig. 9. The linear fit would imply a constant group delay. The second-order fit is the signature of the attochirp. It was discovered later (Kazamias and Balcou, 2004).

dressing laser beam and the harmonics made the sidebands easily observable. The intensity of the laser beam had to be controlled to limit the nonlinear ionization to two photon, in agreement with the theory of Veniard, Taieb, and Maquet (1996). In that case each harmonic has only one sideband on each side, at even multiples of the driving laser frequency. The phase is determined by taking the harmonics in pairs.

It is then fitted to the cosine function of Eq. (1). The "correction" was calculated by Toma and Muller (2002) [Table I in Paul *et al.* (2001)]. The phases versus order extracted from the measurement are shown in Fig. 8. Contrary to the prediction of Antoine, L'Huillier, and Lewenstein (1996), they behave almost linearly [a fit of second order was later found to be more accurate by Kazamias and Balcou (2004) in good agreement of the experimental data and the theory of harmonic generation, revealing the attochirp]. This apparent contradiction will be discussed in the following section. Together with the harmonic intensities obtained from the photoelectron energy spectrum these phases determine uniquely the temporal profile of the total field found as a sequence of 250 as pulses, twice per optical cycle of the laser (Fig. 9).

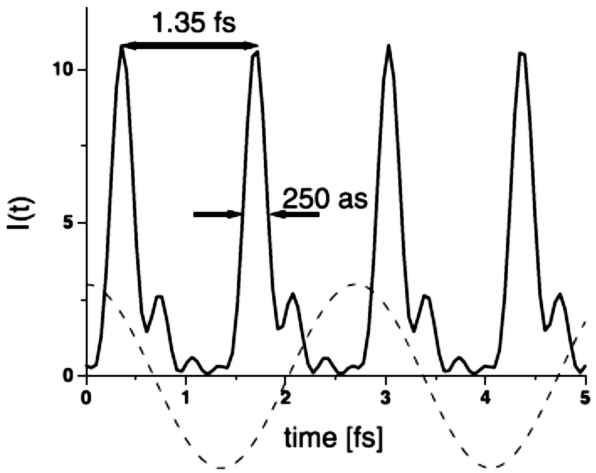

FIG. 9. The first measurement of an APT. From Paul *et al.*, 2001.

The RABBITT method has been used for hundreds of ultrafast experiments (Google Scholar yields 750 results for the search "attosecond RABBITT").

F. The role of the pinhole

The result of Fig. 8 seems to be in flagrant contradiction with the calculation of Fig. 2. However, an experiment by Anne L'Huillier and her group had in fact shed light on the problem (Bellini *et al.*, 1998). In that experiment, harmonic generation is analyzed from the standpoints of coherence and angular distribution. One of the beautiful results is shown in Fig. 2 of the reference, reproduced as Fig. 10. It clearly demonstrates two concentric regions of emission (for harmonic 15 in that case) with different coherence times. What is relevant to the RABBITT experiment is the interpretation in terms of the semiclassical theory of harmonics. The two amplitudes due to the long and short trajectories are at some distance of the emitting jet and appear to be separated in space (this is well understood now) in an annular and a central region respectively. This is where the pinhole plays its part: on top of blocking the annular part of the driving laser, it isolates the short trajectory component. Now, the presence of the two amplitudes in the calculation of Antoine, L'Huillier, and Lewenstein (1996) leads to an interference which explains the phase jumps in Fig. 2, but the pinhole removes the outer part of the harmonic emission, and hence suppresses the interference, leaving, fortunately, only the on-axis emission of the short trajectory. It is somewhat surprising that the long trajectory amplitude, which should dominate the emission, since the field strength which drives it is closer to the field peak and is larger. It is its larger divergence which lowers the efficacy of the long trajectory compared to the *a priori* smaller, short trajectory. Anyway, the pinhole reverses the natural order and practically eliminates the long trajectory. In fact, although a pinhole is not always present in the following setups the emission on-axis is largely favored, if only by the distance between the emitting jet and the location where the APT is used, and the phase-matching condition. The angular distribution of the long trajectory amplitude makes it hard to use it practically despite its theoretical predominance. The case of the production of the first APT was lucky, the interference in the calculation of Antoine, L'Huillier, and Lewenstein (1996) would certainly have been catastrophic. More generally, let's note that the interference long/short has another consequence visible in Lewenstein's quantum theory of harmonic generation: the laser intensity dependence of harmonics, for intensities large enough for the harmonic to be in the plateau region, oscillates strongly due to the interference which was well known ever since 1994 although its effect on the time profile of the emission was not considered until 1996.

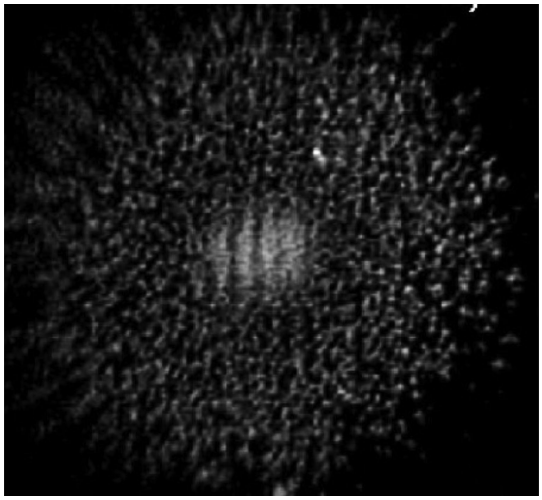

FIG. 10. The long and short trajectories amplitudes correspond to the outer and central parts of the picture respectively. The pinhole eliminated the long trajectory. From Bellini *et al.*, 1998.

The generation of APTs is usually expensive because it needs soft x-rays, vacuum, etc. It is amusing that a cheap pinhole element was the key to the success or failure of the RABBITT method. In that order of ideas, one could wonder how far it would be to go to even shorter pulses. If one starts with a laser on longer wavelength in the near infrared (it seems that a wavelength of 10,000 nm is already available, or will be soon) an obvious advantage, for a given intensity, would be the extension of the harmonic cutoff, and hence of the bandwidth available for shorter pulses. For an intensity of 10^{14} W cm^{-2}, the harmonic cutoff would be pushed to the keV range and pulses in the zeptosecond range. However, the shortest possible pulse is not obtained by using more harmonics, since each harmonic is delayed by a time depending on the order (Mairesse *et al.*, 2003). As is well known the attochirp (5–10 as per order) is difficult to compensate. Within that limit though, the standard setup for an ATP generation is basically the same as for high harmonic generation: either the free propagation on-axis or with the help of a pinhole will guarantee a well-behaved spectral phase.

G. RABBITT and beyond

From 2001, two techniques were shown to generate attosecond pulses. The one described above is very simple and requires only what is necessary to generate high harmonics. In particular, "long" driving pulses (50–70 fs) are permitted. [For more complete characterization see López-Martens *et al.* (2005).] A much more demanding technique was developed in the group of Ferenc Krausz, at that time in Vienna, based on a very short pump pulse but allowing the generation of an isolated attosecond pulse instead of the APT (Hentschel *et al.*, 2001). For measurements of IAP, see, for instance, Kitzler *et al.* (2003).

The price of the simplicity of the first technique is in the temporal profile of the APT, which, by definition, is a periodic sequence of attosecond pulses separated by half the laser period. As for the first mode-locked lasers, before the generalization of the Pockels cell, the temporal resolution was not easy to assert. With the birth of the subcycle physics initiated by Corkum (1993) and Kulander, Schafer, and Krause (1993), it became clear that the periodicity of the train was not an obstacle to study three-step processes, shorter than half a cycle. The number of publications using RABBITT and the APT has been continuously growing since the beginning of the century. Among the most popular applications are the photoionization delays (Dahlström, L'Huillier, and Maquet, 2012) but many other domains from atoms to

FIG. 11. From left: Philippe Balcou, Alfred Maquet, Pierre-Mary Paul, P. A., H. G. Muller, Elena Toma.

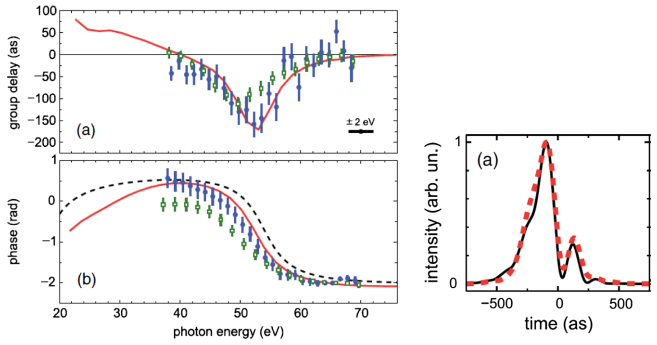

FIG. 12. Left panels: group delay and phase (radians) from RABBITT. Right panel: the time profile of the attosecond pulse is strongly modified by the Copper minimum. From Schoun *et al.*, 2014.

large molecules, clusters to nanostructures to bulk solid, biomatter to condensed phase systems have been investigated (Kitzler and Gräfe, 2016). It is however with IAP that the world record stands with, for example, the ultrabroad band (∼100 eV) pulse engineered in the soft-x-ray domain by Jacob Wörner's group (Gaumnitz *et al.*, 2017), of 43 ± 1 as with the promise eventually of pulses below the atomic unit of 24 as. Other attosecond metrology methods, completing RABBITT and the FROG of Trebino (2000), like CRAB, FROG-CRAB, PROOF, etc. have been described elsewhere (Agostini, Piper, and DiMauro, 2021).

The RABBITT experiment is now over 20 years old. It is perhaps appropriate to cite here the names of the people who accomplished it (Fig. 11): first, I will recall my long-time colleague Pierre Breger and the laser group of the Laboratoire d'Optique Appliquée, Geneviève Mullot, and Frédirika Augé, then the two Ph.D. students Pierre-Marie Paul and Elena Toma, who had a bright career since then, Philippe Balcou who discovered a few years later the second-order fit of the data and the signature of the attochirp (Kazamias and Balcou, 2004), and finally H. G. Muller who is, among heaps of other reasons, the father of the method and author of the acronym RABBITT (Muller, 2002).

The breaking of the femtosecond barrier 20 years ago opened the road to attosecond pulses and subfemtosecond dynamics. Zeptoseconds, the difference of two photoionization delays, so far, are around the corner (Grundmann *et al.*, 2020). While high harmonics are the standard method to produce attosecond pulse trains or isolated attosecond pulses, they suffer from a lack of intensity.

III. SECOND PART: RABBITT AT OHIO STATE

A. Introduction

In this part I would like to briefly review two applications of the APT. The first one is on the measurement of the recombination matrix element near a Cooper minimum (Schoun *et al.*, 2014). The second is in the timing of strong field double ionization (Piper *et al.*, 2024). Both experiments have been carried out at Ohio State.

B. Pulse splitting near a Cooper minimum

As has been well known since John Cooper's paper in 1962, the $p \rightarrow d$ transition matrix element in argon ionization goes through a zero at an energy of about 55 eV. Correspondingly, the phase of the recombination matrix element would undergo a phase jump of π at that energy. Could we access this phase through a RABBITT measurement? And then what are its consequences on the attosecond pulse's shape? If the harmonic is close to the Cooper minimum, the spectral amplitude goes through a minimum while the spectral phase, extracted from RABBITT, is found to make a transition of 2 rad in the same spectral range (close to the expected π jump, due to the contribution of the small $p \rightarrow s$ transition). We showed that the effect of the Cooper minimum is to completely reshape the pulse (Fig. 12).

C. Timing NSDI

The second application of the APT is on the timing of double ionization in the strong field regime, a long-time imagined experiment, finally recently realized at Ohio State.

In HHG, the strong field approximation predicts the value of the cutoff energy and the existence of two main amplitudes for a given harmonic order (Fig. 4). The double ionization (DI) process was explained by a similar theory. In noble gas atoms, the ionization potential of the single charged ion is about twice that of the neutral atoms. From a perturbative point of view, at the same laser intensity, DI would have a much lower probability than single ionization. The experiment [see, for example, Walker *et al.* (1994)] showed that this prediction was off by several orders of magnitude (Fig. 13). The "non-sequential DI" (NSDI) was ascribed (Corkum, 1993) to a similar process as HHG, namely the sequence of tunnel ionization, acceleration in the field and recollision giving rise to an e-2e ionization. The simple process may be complicated by a possible collisional excitation involving states below the DI threshold. In this lecture second part I would like to look at the strong field interpretation of the NSDI from a timing standpoint. I will show that, for the first time, it is possible to clock the process with attosecond precision by selecting the initial ionization time with respect to the optical cycle of the laser which drives the harmonics.

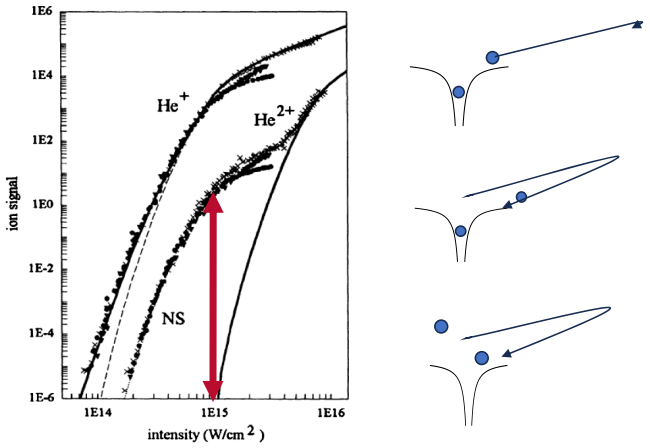

FIG. 13. Left panel the nonsequential double ionization (NS on the figure) is orders of magnitude larger that the sequential process. From Walker *et al.*, 1994. Right panel: the three-step model double ionization (from top: ionization/acceleration, return to the core, and e-2e ejection of a second electron).

This operation has been dubbed "quantum trajectory selector" or QTS (Piper *et al.*, 2024).

D. The QTS experiment

The principle is to start with an attosecond pulse train like those described in the first part. The spectral envelope of the attosecond radiation is sculpted to mimic tunnel ionization, i.e., to produce photoelectrons with essentially zero momentum, by transmission through a thin metallic foil and reflection from a dielectric multilayer mirror. Single ionization of a target atomic jet of argon is thus produced by single photons of the APT in the presence of a strong laser beam and will induce the recollision discussed above (Fig. 13). A pair of glass wedges allow the APT to be delayed with respect to the laser optical cycle. DI of argon produced by the recollision at a controlled phase of the optical cycle is detected in an ion mass/charge time-of-flight spectrometer and monitored as a function of the delay. A typical example is shown Fig. 13. I will not discuss here the details of the recollision model that gives rise to the red curve. One point can be made, though, about the origin of time ($t = 0$). This time is determined by the electrons that do not recollide [see Piper *et al.* (2024)]. Another point is about the observed offset. It is important to realize that although the recollision mechanism of DI is necessary to understand the order of magnitude of the probability, some DI is still due to other processes (Piper *et al.*, 2024). Our results demonstrate control of strong field recollision in the time domain with attosecond precision. Thanks to the APT, we can control and clock the NSDI for the first time (Fig. 14).

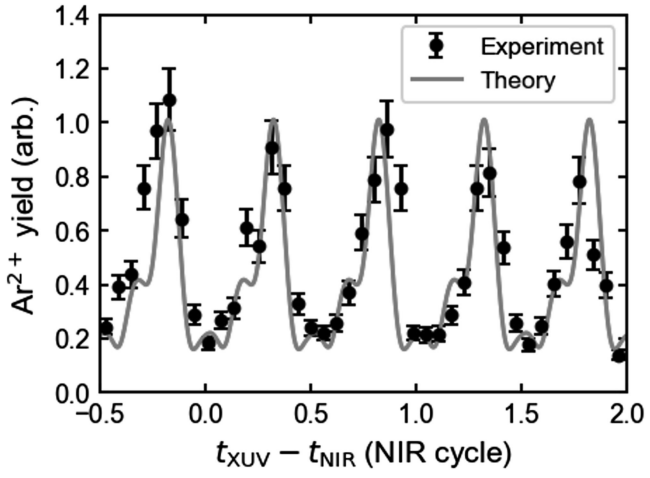

FIG. 14. Time dependence of the double ionization signal with half-cycle period and attosecond duration. The solid line is calculated. From Piper *et al.*, 2024.

IV. CONCLUSION

RABBITT has become the standard method to measure the harmonic phases and to extract the atomic phases. If the phase is a quadratic function of frequency (attochirp), $d\phi/d\omega$ is linear and interpreted as the emission time of the harmonic photons. In the case of a nonlinear dependence of the group delay, as in the vicinity of the Cooper minimum, the pulse profile is deeply modified, and interpretation loses its simplicity.

Over the last 20 years, the measured group delay has proved to be in excellent agreement with the classical dynamics of a free electron in the laser field.

The attochirp/time delay between harmonics is correctly predicted by the model, and imposes a limit to the pulses of APT that one can hope to produce by the high harmonics [see, for example, Mairesse *et al.* (2003) and Doumy *et al.* (2009)]. RABBITT and the ATP have played a major role in the determination of the photoionization delays even though the shortest of attosecond pulses has been recorded with the IAP. The future of attosecond science is perhaps in still another source: the generation of soft x-ray isolated attosecond pulses from an x-ray free-electron laser (FEL) with peak powers larger than 100 GW and durations between 200 and 500 as. The large increase in peak power may allow nonlinear processes, with have been denied so far to high harmonics ATP. Besides, the photon energy of the FEL is much higher (around 600 eV) and could open the road to single particle imaging (Duris *et al.*, 2020). Meanwhile, applications of attosecond pulses in atomic, molecular, and solid-state physics have been extended recently to more complex mediums including large polyatomic molecules, clusters, nanoparticles etc. (Kitzler and Gräfe, 2016).

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